

# PATENT SPECIFICATION

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(54) LIGHT METAL MATRIX COMPOSITE MATERIALS REINFORCED SILICON CARBIDE FIBERS AND A METHOD FOR PRODUCING SAID COMPOSITE MATERIALS

(71) I, HIROSHI WATANABE, President of THE RESEACH INSTITUTE FOR IRON, STEEL AND OTHER METALS OF THE TOHOKU UNIVERSITY of 1-1, Katahira, 2-Chome, Sendai City, Japan, a Japanese subject, do hereby declare the invention, for which I pray that a patent may be granted to me and the method by which it is to be performed, to be particularly described in and by the following statement:

The present invention relates to light metal matrix composite materials reinforced with silicon carbide fibers and a method for producing said composite materials, more particularly aluminum, aluminum alloy or magnesium alloy composite materials reinforced with continuous silicon carbide fibers.

Aluminum is light weight and has an excellent corrosion resistance and a high electric conductivity and therefore has various excellent properties as metal. When copper, magnesium, silicon, manganese, zinc, nickel and the like are alone or together added to aluminum, the mechanical properties are noticeably improved and, in addition to the excellent properties of pure aluminum, the strength is considerably increased at room temperature and at high temperature and the properties, such as a high corrosion resistance, a high abrasion resistance, a low thermal expansion coefficient and the like are added thereto and excellent industrial and engineering materials satisfying each object can be obtained. These materials are used in a broad field of domestic articles, aircraft, automobiles, railway vehicles, ships, architecture and the like and the use amount has been yearly increased.

Magnesium has a specific gravity of 1.74 and is one of the lightest of industrial and engineering metal materials and therefore has been noticed mainly as an aircraft material. The mechanical properties of magnesium alloys are not inferior to those of aluminum alloys and further the machinability is very good but heretofore the industrial purity of the base metal has been very poor in corrosion and this is the greatest drawback. In order to improve the corrosion resistance, various rust preventing surface treatments have been attempted but it has been impossible to overcome this drawback. When magnesium alloys are used as a casting alloy, cracks are caused in crystal grain boundary due to micro-shrinkage or suboxides are admixed owing to oxidation of molten metal and therefore it is difficult to make sound and reliable products and further, since magnesium has a hexagonal close-packed structure, when magnesium alloys are used as forging material, the cold workability is poor compared with that of aluminum and copper which have a cubic crystal structure and therefore the demand for magnesium and magnesium alloys has been small until a recent date.

Since the second world war the speed of aircraft has become higher and jet engines have been rapidly developed. Since magnesium is light weight the development of magnesium as a heat resistant light alloy has been observed and studied extensively. Investigation has been made with respect to the corrosion resistance which is a drawback of magnesium and recently the quality of the base metal has been noticeably improved by an improved process for refining magnesium, so that the corrosion resistance of magnesium has become very high and the crystal grains have been refined by adding zirconium and the like and it has

been possible to produce sound castings, pressure resistant. Furthermore, by addition of materials such as cerium, Misch metal and thorium, heat resistant magnesium alloys having excellent creep properties at high temperature have been produced and various properties of magnesium alloys have been remarkably improved.

5 Furthermore, it has been proposed that the inherent hexagonal close-packed structure of magnesium may be changed into a cubic crystal structure by addition of lithium to provide cold workability.

Heretofore, the study of composite materials consisting of silicon carbide fibers and aluminum, aluminum alloys or magnesium alloys has been made with respect to silicon carbide whiskers and aluminum, aluminum alloys or magnesium alloys. All available silicon carbide fibers have been in the form of whiskers. However, silicon carbide whiskers composed of SiC alone are poor in wettability to aluminum, aluminum alloys or magnesium alloys and the length of the whiskers is a few mm at the longest, so that it is very difficult to arrange said whiskers regularly and the tensile strength is weak, the Young's modulus is low and the cost is high and therefore the whiskers have not been practically used.

10 An object of the present invention is to obviate the above described disadvantages.

The term "continuous silicon carbide fibers" as used herein and in the claims means silicon carbide fibres containing at least 0.01% by weight of free carbon which have been produced by the method described and claimed in British Patent No. 1551342.

15 20 The reason why the silicon carbide fibers containing at least 0.01% by weight of free carbon are used is because the silicon carbide fibers containing less than 0.01% by weight of free carbon are poor in wettability to aluminum, aluminum alloy or magnesium alloy, and even if the composite material is made, when such composite material is subjected to influences of temperature and stress, the mutual reinforcing function cannot be developed, 25 because there are gaps between the fibers and the metal matrix.

In the present invention, silicon carbide fibers containing 0.01-40% by weight of free carbon may be used. The relation of the tensile strength and the Young's modulus of an aluminum composite material containing 25% by volume of the silicon carbide fibers having various amounts of free carbon, to the amount of the free carbon is shown in Fig. 1.

30 35 Until the amount of the free carbon in the silicon carbide fibers is 5% by weight, the tensile strength of the composite material increases and when said amount is from 5% by weight to 20% by weight, the tensile strength gradually lowers. When the amount of the free carbon exceeds 20% by weight, the tensile strength of the composite material decreases greatly.

35 The Young's modulus of the composite material does not substantially vary over a free carbon of 1% by weight.

The reason why the tensile strength of aluminum or aluminum alloy composite materials reinforced with silicon carbide fibers containing free carbon according to the present invention is larger than that of the case where silicon carbide fibers not containing free carbon are used, is presumably based on the fact that the free carbon contained in the silicon carbide fibers reacts with aluminum metal to form aluminum carbide as shown by the following chemical reaction formula (1), whereby chemical adhesion is obtained rather than the physical adhesion.



45 In the case of magnesium alloy composite materials, the free carbon in the silicon carbide fibers also reacts with the elements added to the magnesium alloy, to form carbides of the alloying elements at the surface of the silicon carbide fibers, whereby the chemical adhesion is presumably obtained rather than the physical adhesion.

50 55 In the above described reaction of the free carbon with aluminum or an element added to a magnesium alloy, carbon diffuses from the inner portion of the silicon carbide fiber containing the free carbon to the surface and reacts with aluminum or an element added to a magnesium alloy and further; the aluminum or an element added to a magnesium alloy diffuses into the inner portion of the silicon carbide fiber and reacts with the free carbon, so that the wettability of the silicon carbide fiber and aluminum or an element added in a magnesium alloy becomes very good. The reaction of the free carbon with aluminum or an element added in a magnesium alloy is rapid but the diffusion rates in both directions are slow, so that it is generally advantageous that the fused metal and the silicon carbide fiber containing the free carbon are contacted and reacted for more than 10 minutes. The microscopic photograph of the cross-section of the aluminum composite material in Fig. 2 shows that there is no gaps around the fiber of the composite material obtained by such a chemical reaction and it is known that the above described wettability is very high.

60 65 However, when the composite material produced by using silicon carbide fibers containing a large amount of free carbon is used at a relatively high temperature for a long period of time, the free carbon in the silicon carbide fibers reacts with the metal element in the metal matrix which is apt to form a carbide, to form a carbide, whereby the mechanical

strength of the silicon carbide fibers is not only lowered, but also the composition and the mechanical strength of the matrix itself gradually varies and particularly the brittleness increases.

In particular, when the silicon carbide fibers containing more than 5% by weight of the free carbon are used, this tendency appears and when the amount of the free carbon is more than 20% by weight, the adverse affect of the formation reaction of the carbide becomes remarkable and as shown in Fig. 1, the tensile strength of the composite material lowers. It is due to the hardening function of the formed carbide that the Young's modulus does not vary as shown in Fig. 1, even if the amount of the free carbon in the silicon carbide fibers varies.

Accordingly, in the silicon carbide fibers containing a large amount of the free carbon, it is necessary to restrain the formation reaction of the carbide while maintaining the wettability. When the surface of the silicon carbide fiber is coated with a metal or ceramic having a moderate bonding ability to the fiber, said coated fibers can reinforce aluminum, aluminum alloy or magnesium alloy matrix and can restrain the variation of the physical and chemical properties of the matrix which is caused by the diffusion of the free carbon into the above described matrix.

The silicon carbide fibers to be used in the present invention are produced in accordance with Specification No. 1551342 by spinning organosilicon high molecular weight compounds and baking the spun fibers to obtain continuous silicon carbide fibers but in the course of the production, if the step for removing carbon is omitted or controlled, the continuous silicon carbide fibers containing less than about 40% by free carbon can be obtained.

A brief general explanation will now be given of the method for producing the silicon carbide fibers and the properties.

The silicon carbide fibers containing 0.01-40% by weight of free carbon to be used in the present invention can be produced from the organosilicon compounds classified by the following groups (1)-(10).

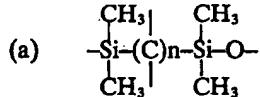
- (1) Compounds having only Si-C bond.
- (2) Compounds having Si-H bond in addition to Si-C bond.
- (3) Compounds having Si-halogen bond.
- (4) Compounds having Si-N bond.
- (5) Compounds having Si-OR bond (R: alkyl or aryl group)
- (6) Compounds having Si-OH bond.
- (7) Compounds having Si-Si bond.
- (8) Compounds having Si-O-SI bond.
- (9) Esters of organosilicon compounds, and
- (10) Peroxides of organosilicon compounds.

At least one of the organosilicon compounds belonging to the above described groups (1)-(10) is subjected to polycondensation reaction by using at least one process of irradiation, heating and addition of polycondensing catalyst to form organosilicon high molecular weight compounds having silicon and carbon as the main skeleton components. For example, the compounds having the following molecular structures are produced.

- (a) -Si-(C)<sub>n</sub>-Si-O-
- (b) -Si-O-(C)<sub>n</sub>-O-
- (c) -Si-(C)<sub>n</sub>-

(d) The compounds having the above described skeleton components (a)-(c) as at least one partial structure in linear, ring and three dimensional structures or a mixtures of the compounds having the above described skeleton components (a)-(c).

The compounds having the above described molecular structures are, for example as follows.



60 n = 1, poly(silmethylenesiloxane),  
n = 2, poly(silethylenesiloxane),  
n = 6, poly(silphenylenesiloxane)

- (b)  $\begin{array}{c} \text{CH}_3 \\ | \\ -\text{Si}-\text{O}-\text{(C)}_n-\text{O}- \\ | \\ \text{CH}_3 \end{array}$
- 5 5
- 10  $n = 1, \text{poly(methyleneoxysiloxane)},$   
 $n = 2, \text{poly(ethyleneoxysiloxane)},$   
 $n = 6, \text{poly(phenyleneoxysiloxane)},$   
 $n = 12, \text{poly(diphenyleneoxysiloxane)}$
- 10 10
- (c)  $\begin{array}{c} \text{CH}_3 \\ | \\ -\text{Si}-\text{(C)}_n \\ | \\ \text{CH}_3 \end{array}$
- 15 15
- 20  $n = 1, \text{polysilmethylene},$   
 $n = 2, \text{polysilethylene},$
- 20 20
- 25 (d) The compounds having the above described skeleton components as at least one partial structure in linear, ring and three dimensional structures or mixtures of the compounds having the above described skeleton components (a)-(c).
- 25 25
- 30 The above described organosilicon high molecular weight compounds are spun and the spun fibers are preliminarily heated under an oxidizing atmosphere and then baked at a high temperature under vacuum or an atmosphere of inert gas, CO gas a hydrogen gas to form silicon carbide fibers having a very high strength and a high Young's modulus.
- 30 30
- 35 The ratio of silicon and carbon contained in the above described organosilicon high molecular weight compounds (a)-(d), which are the starting material of the above described continuous silicon carbide fibers is two silicon atoms to at least five carbon atoms, so that when the organosilicon high molecular weight compounds are spun and the spun fibers are baked, many carbons bonding as the side chain of the high molecular weight compounds volatilize as hydrocarbons or organosilicon compounds but 0.01-40% by weight of carbon remains as the free carbon in the silicon carbide fibers.
- 35 35
- 40 When the content of the free carbon in the silicon carbide fibers is less than 0.01% by weight, the amount of the free carbon which diffuses from the inner portion of the silicon carbide fibers into the metal matrix and reacts with the matrix metal, is too small, so that the tensile strength of the light metal composite materials reinforced with the silicon carbide fibers is not improved. The silicon carbide fibers produced from the organosilicon high molecular weight compounds can not contain more than 40% by weight of free carbon. Accordingly, the content of the free carbon contained in the silicon carbide fibers to be used for the production of the light metal composite materials reinforced with the silicon carbide fibers must be 0.01-40% by weight.
- 40 40
- 45 The tensile strength and the Young's modulus of the silicon carbide fibers to be used in the present invention are shown in the following Table and the silicon carbide fibers having such a high tensile strength have never been known.
- 45 45

Table

50	Tensile strength	300 - 600 kg/mm <sup>2</sup>	50
	Specific strength	about $4.0 \times 10^7$ cm	
	Young's modulus	about 20-40 t/mm <sup>2</sup>	
55	Specific Young's modulus	about $1.10 \times 10^9$ cm	55

- 60 The Young's modulus of the silicon carbide fibers is substantially the same as that of carbon fibers which have the highest Young's modulus among various fibers known presently.
- 60 60
- 65 When a composite material is produced from aluminum or aluminum alloy and the silicon carbide fibers, the free carbon contained in the silicon carbide fibers reacts with aluminum following to the above described formula (1) to form aluminum carbide ( $\text{Al}_4\text{C}_3$ ).
- 65 65
- The standard free energy variation of said formula is negative and the formation of aluminaum carbide is very easy, so that the free carbon on the surface of the silicon carbide fibers reacts with aluminum to form aluminum carbide. This aluminum carbide bonds the

silicon carbide fibers and the matrix of aluminum metal, so that the aluminum carbide acts to improve the wettability of the fibers to aluminum or aluminum alloy.

However, when the silicon carbide fibers contain an amount of the free carbon which exceeds the amount necessary for improving the wettability, the free carbon gradually diffuses into and reacts with aluminum or aluminum alloy to form the carbide in the metal matrix. This carbide lowers the toughness of the metal matrix of the composite material and makes the composite material brittle. Therefore, the tensile strength of aluminum composite materials reinforced with the silicon carbide fibers is lowered.

When an aluminum alloy is used for the matrix, among the elements to be added for formation of aluminum alloys, hafnium, zirconium, titanium, calcium, vanadium, chromium, silicon, manganese, molybdenum, niobium, tantalum and tungsten react with the free carbon in the silicon carbide fibers to form carbides which are stable at a low temperature and further improve the wettability of the silicon carbide fibers to the aluminum alloy. Iron, copper or nickel reacts with SiC, so that the addition of these elements to aluminum or aluminum alloy can improve the wettability of the silicon carbide fibers to aluminum or aluminum alloy. Magnesium alone is scarcely used as an industrial and engineering material and is most used as a magnesium alloy but when a composite material is produced from a magnesium alloy and silicon carbide fibers, the free carbon contained in the silicon carbide fibers reacts with the metals contained in the magnesium alloy which are apt to form a carbide, for example, Al, Mn, Zr, Si, Ca, Y and the other rare earth metals, Th and so on, to form carbides. In this case, when an amount of the carbide formed is small, the formed carbide does not greatly influence the mechanical properties of the metal matrix, but when said amount becomes large, the mechanical properties, particularly toughness of the metal matrix falls and the composite material becomes brittle. Particularly, in the composite materials composed of a heat resistant magnesium alloy and silicon carbide fibers containing more than 5% of the free carbon, during use of the composite materials at a high temperature, the formation of the above described carbides proceeds and when such composite materials are used for a long period of time, the composite materials become gradually brittle.

Even if zinc or beryllium is contained in the magnesium alloy, these elements do not prevent the formation of the carbide with the free carbon in the silicon carbide fibers, so that it is not necessary to consider the influence of these metals.

When the composite materials are produced by using the silicon carbide fibers containing the free carbon which exceeds the amount necessary for improving the wettability of the silicon carbide fibers to the matrix, the surfaces of such silicon carbide fibers are coated with a metal or ceramic in order to prevent the reaction of the free carbon in the silicon carbide fibers with the matrix metal and to maintain the inherent properties of the metal matrix.

In particular, when a composite material produced by using silicon carbide fibers containing 5-40% by weight of the free carbon is used at a high temperature, the mechanical properties of the composite material are lowered due to the formation reaction of the carbide, but when said silicon carbide fibers are coated with a metal or ceramic and a composite material is produced by using such coated fibers as the reinforcing fibers, even if such a composite material is used at a high temperature for a long period of time, the inherent properties of the metal matrix are maintained and the various excellent physical and chemical properties of the above described continuous silicon carbide fibers can be completely developed.

The coating of the above described silicon carbide fibers with a metal, alloy or ceramic can be effected by the following seven processes, namely (1) chemical vapor deposition process, (2) flame fusion spray coating process, (3) sputtering coating process, (4) vacuum evaporating coating process, (5) electroplating process, (6) powder baking process and (7) electroless plating process.

(1) Chemical vapor deposition process:

A metal compound gas alone or together with hydrogen gas, oxygen gas, CO gas or a hydrocarbon gas is thermally decomposed at a temperature range of 500-2700°C to form the above described metal coating on the fiber surface. For example, in the case of tungsten chloride and hydrogen gas, a tungsten metal coating is formed on the fibre about 500°C. When zirconium iodide gas is thermally decomposed at a temperature range of 1300-1800°C, zirconium metal coating is formed on the fibre. When a mixed gas of platinum chloride gas and CO gas is thermally decomposed, platinum coating is formed on the fibre.

(2) Flame fusion spray coating process:

A metal, alloy or ceramics is fused by a flame having a high temperature and the molten metal, alloy or ceramic is sprayed on the silicon carbide fiber surface to form a coating film. Oxygen-acetylene gas flame and a plasma jet flame spraying may be used.

## (3) Sputtering coating process:

Argon plasma is formed by a high frequency discharge under argon atmosphere and said plasma bombards a target of metal, alloy or ceramics to evaporate the metal, alloy or ceramics to form the coating on the silicon carbide fiber surface.

## 5 (4) Vacuum evaporating coating process:

A metal, alloy or ceramic is heated and evaporated in vacuum atmosphere to form the coating on the silicon carbide fiber surface.

## (5) Electroplating process:

10 An electrolyte containing a metal ion is subjected to electrolysis by using the silicon carbide fibers as a cathode to plate the metal coating on the silicon carbide fibers.

## 15 (6) Powder baking process:

Finely divided powders of a metal, alloy or ceramic are prepared and the powders are suspended in a solvent and then the silicon carbide fibers are immersed in the suspension to deposit the powders on the fibers, after which the solvent is evaporated and then heating is effected to form the metal coating on the silicon carbide fibers.

## 15 (7) Electroless plating process:

The silicon carbide fiber surface is plated with a metal without using electric energy. The composition of the plating bath to be used in this process consists of a metal salt, a reducing agent and a buffer solution.

20 Among ceramic coatings, an oxide coating can be formed by six processes excluding the above described process (5).

Other than the above described six processes, it is possible to form an oxide film by heating the silicon carbide fibers or the silicon carbide fibers coated with a metal or alloy at a high temperature. The oxide coating can be formed by heating such fibers at a temperature of 500-2500°C for 0.1-50 hours under an oxidizing atmosphere. At a temperature of lower than 500°C, the oxidizing temperature is low and the oxide coating is not fully formed and at a temperature of higher than 2500°C, the decomposition and evaporation of silicon carbide proceed rapidly, so that the heating temperature must be 500-2500°C. Under an oxidizing atmosphere at a temperature as low as 500°C, a long time of 30 hours is required while at a temperature as high as 2500°C, a good result can be attained in 0.2 hour. As the oxidizing atmosphere for the heating, air is the most economical and the oxide coating can be formed even by using a mixed gas of ozone and air.

25 If the above described heating is effected under at least one gaseous atmosphere of nitrogen gas, ammonia gas and a mixed gas of ammonia gas and hydrogen gas, a metal nitride can be formed and the heating temperature in this case is preferred to be 500-2500°C.

30 When the silicon carbide fiber surface is coated with a metal or alloy, the wettability of the above described metal or alloy to aluminum, aluminum alloy or magnesium alloy matrix is very good, because the contact angle of the mutual metals is small and is less than 90°. Accordingly, the coating of the silicon carbide fiber surface with a metal or alloy improves the wettability of aluminum, aluminum alloy or magnesium alloy matrix to the silicon carbide fibers and further, is effective for preventing the reaction of forming aluminum carbide or carbides with the elements contained in the metal matrix which are apt to form carbides, so that the properties of aluminum, aluminum alloy or magnesium alloy composite materials produced in this manner are noticeably improved. When the metal elements B, Mn, Mo, Al, W, Si, Cr, Ca, V, U, Th, Nb, Ta, Ti, Zr and Hf among the coated metals and alloys are respectively heated together with the free carbon, stable carbides are formed. Accordingly, the above described metal coating is partly or entirely converted into the carbide coating through heating when the composite material is formed and the carbide coating prevents the diffusion of the free carbon from the inner portion of the silicon carbide fibers and the reaction of the free carbon with aluminum or the metal element in magnesium alloy to form the carbide.

35 The advantageous metals to be used for coating the silicon carbide fibers other than the above described metals capable of forming the carbides are Be, Mg, Fe, Co, Ni, Cu, Zn, Ge, Pd, Ag, Cd, Sn, Sb, Pt, Au, and Pb and alloys of at least two of the above described metal elements.

40 The advantageous ceramics to be used for coating the silicon carbide fibers include MgO, Al<sub>2</sub>O<sub>3</sub>, SiO<sub>2</sub>, TiO<sub>2</sub> and ZnO as the oxide and AlN, Mg<sub>3</sub>N<sub>4</sub>, Si<sub>3</sub>N<sub>4</sub>, TiN and ZrN as the nitride and TiC, ZrC and WC as the carbide.

45 When the thickness of the film coated on the above described silicon carbide fibers is less than 100Å, the coated film is too thin and it has little ability to prevent the diffusion of the free carbon while at a thickness of more than 2000Å, its diffusion inhibiting property reaches a limiting value so that no greater thickness is any advantage. Accordingly, the thickness of the film to coat the silicon carbide fibers containing 0.01-40% by weight of the free carbon is preferred to be 100-2000Å.

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The composite materials composed of the silicon carbide fibers and aluminum, aluminum alloy or magnesium alloy can be produced by the following methods for producing metal composite materials reinforced with fibers, for example,

- 5      (1) diffusion bonding process,
- (2) liquid infiltration process,
- (3) fusion spray process,
- (4) electrodeposition process,
- (5) hot extrusion and hot rolling process,
- 10     (6) chemical vapor deposition process, and
- (7) cold pressing and sintering process.

10     (1) Diffusion bonding process:  
 The silicon carbide fibers and the matrix metal wires are arranged in alternative directions, the upper surface and the bottom surface of the piled silicon carbide fibers and metal wires are covered with thin films of the matrix metal or only the bottom surface is covered with the above described thin film and the upper surface is covered with matrix metal powders mixed with an organic binder to form composite layers. The said layers are then bound together and the formed laminate is then heated under a pressure to form a composite material composed of the silicon carbide fibers and the matrix metal. As the above described organic binder, substances which volatilize before heating to a temperature at which the matrix metal and the free carbon in the silicon carbide fibers react and form the carbide, are desirable and for example, starch, carboxymethylcellulose, paraffin, resin, ammonium chloride, mineral oil, polyvinyl alcohol, polystyrene, organic polymers and so on can be used.

20     (2) Alternatively, the silicon carbide fibers coated with matrix metal powders are mixed with an organic binder and arranged and piled and the formed laminate is pressed, under heating, to form a composite material.

25     (2) Liquid infiltration process:

30     The spaces between the arranged silicon carbide fibers are filled with molten aluminum, aluminum alloy or magnesium alloy. In this case, since the wettability of the silicon carbide fibers coated with the metal to the matrix metal is good, the spaces between the arranged fibers can be thoroughly filled with the matrix metal.

35     (3) Fusing spray process:

35     The surfaces of the arranged silicon carbide fibers are coated with the matrix metal by a plasma fusion spray or a gas fusion spray to produce a tape-shaped composite material. This tape-shaped composite material is directly used or the tape-shaped composite materials are piled and the formed laminate is subjected to the above described diffusion bonding process (1) to form a composite material.

40     (4) Electrodeposition process:

40     The matrix metal is electrolytically deposited on the surfaces of the fibers to form a composite material and further it is possible to arrange and pile the formed composite materials and the formed laminate is subjected to the above described diffusion bonding process (1).

45     (5) Hot extrusion and rolling process:

45     The silicon carbide fibers are arranged in one direction and said arranged silicon carbide fibers are put between matrix metal foils in a sandwich form and then passed through heated rolls to bond the fibers and the matrix metal, whereby a composite material is produced.

50     (6) Chemical vapor deposition process:

50     The silicon carbide fibers are charged in a heating furnace and, for example, a mixed gas of aluminum chloride and hydrogen gas is introduced therein. The aluminum chloride is thermally decomposed to deposit aluminum metal on the surfaces of the silicon carbide fibers and to form a composite material. Furthermore, the metal deposited fibers may be arranged and piled and the formed laminate is subjected to the above described diffusion bonding process (1).

55     (7) Cold pressing and sintering process:

55     The spaces between the arranged fibers are filled with matrix metal powders and the assembly is molded under pressure, and the formed molding is heated and sintered under pressure or no pressure to form a composite material.

60     When the composite materials are produced by using the silicon carbide fibers coated with the metal or ceramics, the above described seven processes (1)-(7) may be applied.

60     The tensile strength ( $\sigma_c$ ) of the composite material produced from the silicon carbide fibers and the metal matrix is represented by a formula (2).

$$\sigma_c = \sigma_f V_f + \sigma_m V_m \quad \dots (2)$$

$\sigma_c$ : Tensile strength of the composite material.

65      $\sigma_f$ : Tensile strength of SiC fibers.

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$\sigma_m$ : Tensile strength of the metal matrix.

$V_f$ : % by volume of SiC fibers

$V_m$ : % by volume of the metal matrix.

As seen from the above formula (2), the tensile strength of the composite material becomes larger with increase in the volume percent of the silicon carbide fibers in the composite material. Accordingly, production of the composite material having a higher tensile strength needs an increase of the volume percent of the silicon carbide fibers added. However, when the amount of the silicon carbide fibers exceeds 70% by volume, the amount of the metal matrix is too small so that it is impossible to fill fully the spaces between the silicon carbide fibers with the metal matrix, accordingly it is impossible to develop the tensile strength shown by the above formula (2) in the formed composite material. When the amount of the fibers becomes smaller, the tensile strength of the composite material falls, so that not less than 20% by volume of the silicon carbide fibers should be added in order to obtain practically useful composite materials. Accordingly, in the production of light metal composite material reinforced with silicon carbide fibers according to the present invention, the amount of silicon carbide fibers to be added must be 20-70% by volume.

The Young's modulus ( $E_c$ ) of the composite material is shown by the following formula

$$(3) \quad E_c = V_f E_f + V_m E_m \quad \dots(3)$$

$E_c$ : Young's modulus of the composite material.

$E_f$ : Young's modulus of SiC fibers.

$E_m$ : Young's modulus of the metal matrix.

$V_f$ : % by volume of SiC fibers.

$V_m$ : % by volume of the metal matrix.

As seen from the above formula, the Young's modulus of the composite material becomes larger with increase in the amount of the silicon carbide fibers added to the metal matrix. However, when the amount of the fibers becomes too large, the toughness of the composite materials becomes poor, so that the composite materials become brittle and lack reliability.

The present invention will be explained in more detail.

For a better understanding of the invention, reference is made to the accompanying drawings, wherein:

Fig. 1 shows the curves of the relationship between the amount of the free carbon in the silicon carbide fibers to the tensile strength and Young's modulus of one embodiment of the composite material according to the present invention,

Fig. 2 shows a photomicrograph of the cross-section of an aluminum composite material according to the present invention,

Fig. 3 shows a perspective view of the cross-section of the aluminum composite foil reinforced with the silicon carbide fibers obtained in Example 11,

Fig. 4 shows a optical microscope photograph of the cross-section of an aluminum composite material reinforced with the silicon carbide fibers coated with copper, and

Fig. 5 shows the relationship of the tensile strength to the temperature variation of the aluminum composite materials produced by using the silicon carbide fibers coated by various metals.

The following examples are given for the purpose of illustration of this invention and are not intended as limitations thereof.

#### Example 1

Dimethyldichlorosilane and sodium were reacted to produce polydimethylsilane. 250g of polydimethylsilane was charged in an autoclave having a capacity of 1ℓ and air in the autoclave was substituted with argon gas and a reaction was effected at 470°C for 14 hours. After completion of the reaction, the formed polycarbosilane was discharged as a n-hexane solution. This n-hexane solution was filtered to remove impurities and the n-hexane was evaporated under a reduced pressure, after which the residue was concentrated by heating in an oil bath at 280°C under vacuum for 2 hours. Polycarbosilane was obtained in a yield of 40% based on the dimethyldichlorosilane content. The average molecular weight of the formed polycarbosilane was 1700. By using a usual spinning apparatus, the polycarbosilane was heated and melted at 330°C under argon atmosphere to form a spinning melt and was spun at a spinning rate of 200m/min to obtain polycarbosilane fibers. The fibers were heated by raising the temperature from 20°C to 190°C in air in 6 hours and this temperature was kept for 1 hour to prevent cohesion of the fibres. The thus treated fibers were heated to 1300°C at a temperature raising rate of 100°C/hr under vacuum of  $1 \times 10^{-3}$  mmHg and this temperature was kept for 1 hour to form SiC fibers. The formed SiC fibers had an average diameter of  $10\mu\text{m}$ , an average tensile strength of  $350\text{kg/mm}^2$ , an average Young's modulus of  $23 \times 10^3 \text{kg/mm}^2$  and a specific gravity of  $2.70\text{g/cm}^3$ .

5        Aluminum was heated and melted in a vacuum chamber of  $3 \sim 6 \times 10^{-6}$  mmHg at 800°C. A bundle of parallel continuous silicon carbide fibers having a diameter of  $10\mu$  and containing 5% by weight of free carbon was charged in an alumina pipe, one end of which was sealed and this alumina pipe was introduced into the above described vacuum chamber. Then, the open end of the alumina pipe containing the above described fibers was submerged in the previously melted aluminum metal bath and then argon gas was flowed into the vacuum chamber to raise the pressure in the chamber to 1 atm.

10      The molten aluminum was pushed up into the alumina pipe and filled between the continuous silicon carbide fibers and reacted with the free carbon in the fibers to form  $\text{Al}_2\text{C}_3$ . The molten state of the aluminum was maintained for 1 hour to conduct this formation reaction fully and to make the wettability of the continuous silicon carbide fibers and aluminum higher. The amount of the silicon carbide fibers in the aluminum composite material was 41% by volume. The tensile strength of the composite material was 105kg/mm<sup>2</sup>.

15      Since the tensile strength of aluminum casting is about 10kg/mm<sup>2</sup>, the composite material produced by the method of the present invention has a tensile strength of about 10 times that of the aluminum castings.

*Example 2*

20      Aluminum alloy containing 0.3% by weight of silicon and 0.5% by weight of magnesium was heated and melted in a vacuum chamber of  $1 \times 10^{-5}$  mmHg at 705°C and maintained molten.

25      A bundle of the continuous silicon carbide fibers, each fiber containing 4% by weight of free carbon and having a diameter of  $8\mu\text{m}$ , was charged in parallel into an alumina pipe, both ends of which were opened, one end of said pipe was then sealed, the other end of said pipe was connected to a vacuum apparatus, said pipe was put in the vacuum chamber and heated and the continuous silicon carbide fibers were evacuated. Then, the other end of said alumina pipe was submerged in the above described molten aluminum-silicon-magnesium alloy, after which argon gas was passed into the above described vacuum chamber to raise the pressure in the chamber to 5mmHg and the pressure in said alumina pipe was  $1 \times 10^{-5}$  mmHg of vacuum and the molten aluminum-silicon-magnesium alloy was sucked up in the alumina pipe housing the continuous silicon carbide fibers. The aluminum alloy was maintained molten for 40 minutes to react the free carbon in said silicon carbide fibers with the aluminum and silicon of the alloy to form an aluminum alloy composite material reinforced with continuous silicon carbide fibers. The amount of silicon carbide fibers in the above described composite material was 48% by volume and the tensile strength of the composite material was 130kg/mm<sup>2</sup>.

*Example 3*

30      In the same manner as described in Example 2, a molten aluminum alloy containing 4.5% by weight of copper was contacted with the silicon carbide fibers containing 3% by weight of free carbon for 30 minutes to make the wettability of the above described fibers to aluminum-copper alloy higher, whereby an aluminum alloy composite material reinforced with silicon carbide fibers was obtained which contained 28% by volume of silicon carbide fibers and had a tensile strength of 86kg/mm<sup>2</sup>.

*Example 4*

35      In the same manner as described in Example 2, a molten aluminum alloy containing 0.3% by weight of silicon, 0.6% by weight of iron and 5% by weight of copper was contacted with the silicon carbide fibers containing 5.5% by weight of free carbon for 20 minutes to make the wettability of the silicon carbide fibers to the above described aluminum alloy higher, whereby an aluminum alloy composite material reinforced with the silicon carbide fibers was obtained which contained 34% by volume of silicon carbide fibers and had a tensile strength of 103kg/mm<sup>2</sup>.

*Example 5*

40      A net having about 250 meshes, which was obtained by knitting silicon carbide fibers having a diameter of  $10\text{-}15\mu\text{m}$  and containing 3% by weight of free carbon, and aluminum foils were piled alternately and subjected to hot pressing by applying a pressure of 5 tons/cm<sup>2</sup> under vacuum at a temperature of 600°C for 40 minutes to obtain an aluminum composite material reinforced with the silicon carbide fibers which contained 21% by volume of silicon carbide fibers and had a tensile strength of 54kg/cm<sup>2</sup>.

45      A continuous carbide fiber having a diameter of  $10\mu\text{m}$  and containing 6% by weight of free carbon was passed through a bath having a length of 1m and containing molten aluminum heated at 800°C under argon atmosphere at a feed rate of 8cm/min to make the wettability of the continuous silicon carbide fiber to aluminum higher, whereby an aluminum composite wire reinforced with the continuous silicon carbide fiber was obtained. The formed wire had a diameter of  $30\mu\text{m}$ , the tensile strength was 51kg/mm<sup>2</sup>, which is about 5 times as high as that of aluminum and the Young's modulus was  $15.0 \times 10^3$  kg/mm<sup>2</sup>, which is about 2 times as high as that of aluminum.

*Example 7*

5 A magnesium alloy consisting of 10% by weight of aluminum, 0.5% by weight of manganese and the remainder magnesium was heated and melted in a chamber under an argon atmosphere at 1050°C. A bundle of parallelly arranged silicon carbide fibers, each having a diameter of 10 $\mu\text{m}$  and containing 4% by weight of free carbon was charged into a magnesia pipe, both ends of which were opened, and one end of said pipe was sealed and the other end was connected to a vacuum system and the pipe was evacuated while heating and said pipe was put in the above described chamber under argon atmosphere and the sealing was removed. The opened end of this magnesia pipe was submerged in the molten magnesium alloy and the other end was made evacuated. The molten magnesium alloy was pushed up into the magnesia pipe and filled between the silicon carbide fibers and the magnesium alloy was maintained molten for 30 minutes to obtain a magnesium alloy composite material reinforced with the silicon carbide fibers which contained 25% by volume of the fibers and had a tensile strength of 73kg/mm<sup>2</sup>. This tensile strength was about 4 times as high as that of the magnesium alloy containing no silicon carbide fibers. From this result, it can be seen that the reinforcing effect of the silicon carbide fibers is fully developed and the free carbon in the silicon carbide fibers reacts with the aluminum in the magnesium alloy and the wettability of the fiber to the metal matrix is good.

*Example 8*

10 A magnesium alloy consisting of 93.4% by weight of magnesium, 0.6% by weight of zirconium, 2.0% by weight of yttrium and 4.0% by weight of zinc was heated and melted in a chamber under argon gas atmosphere at 800°C. A bundle of parallelly arranged silicon carbide fibers, each having a diameter of 15 $\mu\text{m}$  and containing 6% by weight of free carbon was charged in a magnesium pipe, both ends of which were opened and in the same manner as described in Example 7, the spaces between the silicon carbide fibers were filled with the fused magnesium alloy which was kept molten for 1 hour to obtain a magnesium alloy composite material reinforced with the silicon carbide fibers. The composite material contained 32% by volume of the silicon carbide fibers and had a tensile strength of 87kg/mm<sup>2</sup>.

*Example 9*

15 Silicon carbide fibers, containing 5% by weight of free carbon, were piled in parallel and the spaces between the silicon carbide fibers were filled with magnesium alloy powders consisting of 1.0% by weight of manganese, 0.1% by weight of calcium, 0.25% by weight of silicon, 0.03% by weight of copper, 0.008% by weight of nickel, 0.20% by weight of other minor trace elements and 98.412% by weight of magnesium to form a molding. Said molding was subjected to hot pressing under a pressure of 0.5ton/cm<sup>2</sup> in an argon atmosphere at 550°C for 4 hours to obtain a magnesium alloy composite material reinforced with silicon carbide fibers. The composite material contained 30% by volume of the fibers and had a tensile strength of 30kg/mm<sup>2</sup>. This tensile strength was about twice as high as that of the magnesium alloy containing no fibers.

*Example 10*

20 A woven fabric composed of silicon carbide fibers, each having a diameter of 10 $\mu\text{m}$  and containing 10% by weight of free carbon was cut into discs having a diameter of 100mm. The formed discs were arranged at a distance of 0.02mm and such arranged discs were put in a chamber under argon atmosphere. A magnesium alloy, consisting of 9.5% by weight of aluminum, 0.5% by weight of manganese, 2.1% by weight of zinc, 0.2% by weight of silicon, 0.1% by weight of copper, 0.05% by weight of nickel, 0.25% by weight of trace elements and 87.3% by weight of magnesium was charged in the chamber under an argon atmosphere and melted by heating at 800°C and the molten alloy was poured onto the above described arranged woven fibrous fabrics. The magnesium alloy was kept molten for 30 minutes to obtain a magnesium alloy composite material reinforced with the silicon carbide fibers. The composite material contained 25% by volume of the silicon carbide fibers and had a tensile strength of 65kg/mm<sup>2</sup>. This tensile strength was about 3 times as high as that of the magnesium alloy containing no fibers.

*Example 11*

25 The surfaces of silicon carbide fibers containing 13% by weight of free carbon (average diameter: 20 $\mu\text{m}$ , average tensile strength: 300kg/mm<sup>2</sup>, average Young's modulus: 3.0x10<sup>4</sup>kg/mm<sup>3</sup>) were coated with a vacuum evaporating coating process to form a coating film having a thickness of 400-500Å of tungsten, molybdenum, copper, silicon, magnesium, zinc, iron, manganese, nickel, titanium or silver metal.

30 The coated silicon carbide fibers were arranged in parallel in a layer on a pure aluminum foil having a thickness of 0.05mm and then the silicon carbide fiber layer was covered with pure aluminum foil and the assembly was passed through hot rolls heated at a temperature of 600°C to form a composite foil consisting of the silicon carbide fibers and aluminum. The schematic view of this composite foil is shown in Fig. 3. 20 composite foils were superposed and subjected to hot pressing at a temperature of 600°C to obtain an aluminum composite

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material reinforced with the silicon carbide fibers.

The composite material contained 25% by volume of silicon carbide fibers. As one embodiment, when the structure of the composite material was observed with optical microscope, a structure photograph of the composite material manufactured by using the silicon carbide fibers coated with copper is shown in Fig. 4. As shown in Fig. 4, the bonding of the silicon carbide fibers and aluminum is very good and the silicon carbide fibers tightly adhere to aluminum. The mechanical properties of the composite materials produced by using the silicon carbide fibers coated with the metals as shown in the following Table 1 are shown in the following Table.

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Table 1

	Coating metal element											15
	W	Mo	Cu	Si	Mg	Zn	Fe	Mn	Ni	Ti	Ag	
	Tensile strength (kg/mm <sup>2</sup> )	63	51	72	49	48	56	68	60	80	70	73
	Young's modulus (10 <sup>3</sup> kg/mm <sup>2</sup> )	16	15	14	16	13	15	17	16	14	16	13
	Elongation (%)	2.3	2.4	2.1	2.8	3.1	3.3	2.3	2.7	3.0	2.6	3.4

The tensile strength of pure aluminum is about 11kg/mm<sup>2</sup>, its Young's modulus is about 7x10<sup>3</sup>kg/mm<sup>2</sup> and its elongation is about 25%. The tensile strengths of the composite materials shown in the above Table 1 more or less vary depending upon the coating metal but are a few times as high as that of pure aluminum. Young's moduli of the composite materials are about twice as high as that of the pure aluminum. The elongations of the composite materials are far smaller than that of pure aluminum. However, in the composite material produced from the silicon carbide fibers not coated with metal and pure aluminum, the elongation is less than 2% and is very small. When compared with this value it can be seen that the effect of coating is noticeable. The relationship of the tensile strengths of the above described composite materials with temperature is shown in Fig. 5. As seen from Fig. 5, pure aluminum is low in tensile strength even at room temperature and when the temperature exceeds 200°C, the tensile strength falls further. However, the composite materials of the present invention only show a small decrease in tensile strength at up to 500°C and maintain a fairly high strength, and can be used at a high temperature. Furthermore, even when these composite materials were kept at a high temperature of 400°C for 150 hours hardly any variation of properties was observed.

#### Example 12

The surfaces of silicon carbide fibers containing 20% by weight of free carbon, (tensile strength: 320kg/mm<sup>2</sup>, Young's modulus: 2.8x10<sup>4</sup>kg/mm<sup>2</sup>) were separately coated with 18 Cr stainless steel, Inconel X (Trade Mark) ASTM 1A copper alloy, Zircalloy 2 and Ti-8Mn titanium alloy by means of a usual flame fusion spray coating apparatus to form each coating film having a thickness of about 600Å.

The above described coated silicon carbide fibers were arranged in parallel and charged in a stainless foil mold of 2mmx2mmx100mm and one end of said mold was put in molten aluminum (1060-H 18 aluminum alloy) heated at 800°C and pressure in said mold was reduced from the other end, whereby the fused aluminum was sucked up into the coated silicon carbide fibers. Then, the mold was cooled, after which the stainless foil was broken and an aluminum composite material was taken out. The aluminum composite materials obtained had 40% by volume of silicon carbide fibers. The mechanical properties of these composite materials at room temperature are shown in the following Table 2.

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Table 2

		Coating alloy					5
		18 Cr stainless	Inconel X	Zircalloy 2	Ti-8Mn	ASTM1A copper alloy	
10	Tensile strength ( $\text{kg}/\text{mm}^2$ )	125	141	138	115	121	10
15	Young's modulus ( $10^3 \text{kg}/\text{mm}^2$ )	21	22	21	18	19	15
	Elongation (%)	2.6	2.9	2.4	3.1	2.8	

20 The tensile strength of the matrix of 1060-H 18 aluminum alloy is about  $13\text{kg}/\text{mm}^2$  and the Young's modulus is about  $9 \times 10^3 \text{kg}/\text{mm}^2$  and the elongation is about 6%. As seen from the above Table 2, in the aluminum composite materials according to the present invention, the tensile strength is about 10 times as high as that of the matrix, the Young's modulus is about 2.5 times as high as that of the matrix and the elongation is about 3% and these data show that the aluminum composite materials of the present invention have excellent properties. Furthermore, even when the composite materials were kept at a high temperature of 450°C for a long time (150 hours), the properties did not change.

25 Example 13  
30 The surfaces of silicon carbide fibers containing 5% by weight of free carbon (tensile strength:  $340\text{kg}/\text{mm}^2$ , Young's modulus:  $35 \times 10^4 \text{kg}/\text{mm}^2$ ), Young's modulus:  $35 \times 10^4 \text{kg}/\text{mm}^2$ ) were separately coated with ZrN, CaO, TiC, HfB and Nb<sub>5</sub>Si<sub>3</sub> to a thickness of about 800Å under argon atmosphere by means of a high frequency bipolar sputtering apparatus. The coated silicon fibers were arranged in parallel in a layer on an aluminum foil having a thickness of 0.2μm and such arranged silicon carbide fiber layer was covered with fine aluminum powder (5154-0 aluminum alloy) or less than 325 mesh and the assembly was subjected to hot pressing under a pressure of 200kg/cm<sup>2</sup> at a temperature of 560°C to obtain a composite foil. 20 composite foils were superposed and subjected to hot pressing under the same conditions as described above to obtain aluminum composite materials. The composite materials obtained contained 30% by volume of the silicon carbide fibers. The mechanical strengths of the composite materials are shown in the following Table 3.

Table 3

45		Ceramic coating					45
		ZrN	CaO	TiC	HfB	Nb <sub>5</sub> Si <sub>3</sub>	
50	Tensile strength ( $\text{kg}/\text{mm}^2$ )	104	98	89	110	101	50
	Young's modulus ( $10^3 \text{kg}/\text{mm}^2$ )	23	27	16	21	25	
	Elongation (%)	3.1	2.9	3.3	2.5	2.2	

55 The tensile strength of 5140-0 aluminum alloy is 24 kg/mm<sup>2</sup> and the elongation is 27%. As shown in the above Table 3, the tensile strengths of the composite materials according to the present invention are very high and the Young's moduli are also high and the elongation is about 3% and these materials can be satisfactorily used as the composite material. Even when these composite materials were kept at 450°C for a long time (150 hours), the properties did not change.

60 Example 14  
65 Silicon carbide fibers containing 10% by weight of free carbon (tensile strength:  $300\text{kg}/\text{mm}^2$ , Young's modulus:  $2.5 \times 10^4 \text{kg}/\text{mm}^2$ ) were coated with Al, Zr, Cr, or V metal to a thickness of 500-1000Å by means of a vacuum evaporating coating apparatus. Then,

the silicon carbide fibers coated with  $\text{Al}$  were oxidized at 500°C for 1 hour to form silicon carbide fibers coated with  $\text{Al}_2\text{O}_3$ . The other metal coatings were oxidized at 1000°C in air to form the silicon carbide fibers coated with  $\text{ZrO}_2$ ,  $\text{Cr}_2\text{O}_3$  or  $\text{V}_2\text{O}_5$ .

The above described coated silicon carbide fibers were coated with aluminum alloy (5357-H38) to a thickness of 0.1-10 $\mu\text{m}$  by means of a flame fusion spray coating apparatus. The silicon carbide fibers coated with the aluminum alloy were piled in a mold of 7mmx10mmx50mm and subjected to hot pressing under a pressure of 200kg/mm<sup>2</sup> at 500°C to obtain aluminum composite materials. These composite materials contained 32% by volume of the silicon carbide fibers. The mechanical properties of the composite materials are shown in the following Table 4.

Table 4

		Ceramic coatings				15
		$\text{Al}_2\text{O}_3$	$\text{ZrO}_2$	$\text{Cr}_2\text{O}_3$	$\text{V}_2\text{O}_5$	
15	Tensile strength (kg/mm <sup>2</sup> )	87	93	78	96	
20	Young's modulus (10 <sup>3</sup> kg/mm <sup>2</sup> )	21	18	19	25	20
25	Elongation (%)	2.3	3.1	2.1	2.5	

The tensile strength of the aluminum alloy (5357-H38) was 23kg/mm<sup>2</sup>. The tensile strengths of the composite materials according to the present invention are 4-5 times as high as that of the above described aluminum alloy and composite materials having excellent properties were produced. Even when these composite materials were kept at a high temperature of 450°C for a long time (150 hours), the properties did not change.

#### Example 15

Silicon carbide fibers containing 13% by weight of free carbon were baked at 1200°C in air for 2 hours to form  $\text{SiO}_2$  coating films on the surfaces of the fibers and simultaneously to remove free carbon. The silicon carbide fibers coated with  $\text{SiO}_2$  were closely arranged in a layer on a pure aluminum foil having a thickness of 0.05 mm and then the silicon carbide fiber layer was covered with more pure aluminum foil having the same thickness as described above and the assembly was subjected to hot rolling while heating at 650°C to form a composite foil. 20 composite foils were superposed and subjected to hot pressing at a temperature of 640°C for 1 hour to obtain an aluminum composite material reinforced with silicon carbide fibers. The aluminum composite material obtained contained 25% by volume of the silicon carbide fibers. The tensile strength of the composite material was 110kg/mm<sup>2</sup>, while the tensile strength of the composite material produced from the silicon carbide fibers not coated with  $\text{SiO}_2$  and aluminum was 35kg/mm<sup>2</sup>. As seen from the comparison, the tensile strength of the composite material according to the present invention is much larger than this comparative composite material. Even when the composite material was kept at 450°C for a long time (100 hours), the properties did not change.

#### Example 16

Magnesium alloy composite materials were produced from 60% by volume of magnesium alloy consisting of 3.0% by weight of aluminum, 1% by weight of manganese, 1.3% by weight of zinc and remainder being magnesium and 40% by volume of the silicon carbide fibers containing 15% by weight of free carbon. Silicon carbide fibers (diameter: 20 $\mu\text{m}$ ) were coated with nickel, copper or 13% iron-chromium alloy to a thickness of about 800Å by a vacuum evaporating coating process.

The silicon carbide fibers having a diameter of 20 $\mu\text{m}$  were heated at 1,100°C in air for 1 hour to obtain the silicon carbide fibers coated with silicon oxide.

The surfaces of these fibers were coated with a mixture of the above described magnesium alloy powders (95% by weight) and paraffin (5% by weight) to an average thickness of 12 $\mu\text{m}$  and the thus treated fibers were arranged and piled in a mold of 20mmx50mmx30mm and kept under a pressure of 200kg/mm<sup>2</sup> at 480°C in argon gas atmosphere for 1 hour to obtain magnesium alloy composite materials. The tensile strengths and the Young's moduli of the composite materials are shown in the following Table 5.

Table 5

		Coating material				5
		Nickel	Copper	13% iron-chromium	SiO <sub>2</sub>	
10	Tensile strength (kg/mm <sup>2</sup> )	95	99	87	85	10
	Young's modulus (10 <sup>3</sup> kg/mm <sup>2</sup> )	16	18	17	16	
15	Elongation (%)	1.5	1.2	1.6	1.8	15

In the magnesium alloy, the tensile strength is 22kg/mm<sup>2</sup> and the Young's modulus is 4.6x10<sup>3</sup>kg/mm<sup>2</sup> and as compared with these values, the tensile strength and the Young's modulus of the magnesium alloy composite materials according to the present invention were much higher as seen from the above Table 5. Even when the magnesium alloy composite materials were kept at 480°C for a long time (50 hours), the properties did not lower.

#### Example 17

Surfaces of the silicon carbide fibers containing 13% by weight of free carbon (average diameter: 15μm, average tensile strength: 250kg/mm<sup>2</sup>, average Young's modulus: 20ton/mm<sup>2</sup>) were plated with copper to a thickness of 0.2-0.4μm by means of a electroless plating liquid.

The above described copper plated silicon carbide fibers were arranged uniformly on an aluminum (HZ/102) foil and aluminum was plasma sprayed on to the fibres and foil to produce a composite foil. The above described composite foils were superposed and subjected to hot pressing under a pressure of 50kg/cm<sup>2</sup> at 630°C in vacuum for 90 minutes to produce a composite material. The composite material obtained contained 30% by volume of silicon carbide fibers and had a tensile strength of 55kg/mm<sup>2</sup>. Even if this composite material was heated at 500°C for a long time, the tensile strength did not change. The aluminum, aluminum alloy or magnesium alloy composite materials reinforced with the continuous silicon carbide fibers obtained in the present invention have a high tensile strength and a high Young's modulus, so that composite materials can be used as in following various applications.

#### (a) Materials for apparatus for producing synthetic fibers:

Bobbin, Separator, Pump parts, Ball, Sleeve, Mechanical seal, Valve, Nozzle, Stirrer, Reaction Vessel, Pipe and Heat exchanger.

#### (b) Materials for apparatus for synthetic chemistry:

Plunger pump, Sleeve, Mechanical seal, Separator, Reactor valve, Reducing valve, Seat, Heat exchanger, Centrifugal machine and Vessel for a low temperature.

#### (c) Mechanical industrial materials:

Heat exchanger, Die for pressing powders, Ultrasonic working machine, Honing machine, Sewing machine parts, Cam, Ball-mill parts, Camera parts, vacuum pump, Collector, Bearing, Tool, Watch parts and machine base.

#### (d) Materials for domestic and office supplies:

Desk, Various shelves, Chair and Various lockers.

#### (e) Materials for constructing machines:

Boring machine, Rock crusher, Crusher, Sand pump, and Power shovel.

#### (f) Fire protecting equipment such as sprinklers.

#### (g) Marine materials:

Heatexchanger, radio Antenna, Buoy on water and Tank.

#### (h) Automobile materials:

Engine, Manifold, carrier for differential gear, Crank case, Pump body, Valve body, Clutch housing, Case for transmission, Gear box, Fly-wheel housing, Cylinder block, Cylinder head, Piston, Pulley, Pump body, Blower housing, Tire mold, Rotary engine, and Body material.

#### (i) Materials for apparatus for producing food:

Valve, processing vessel, Mechanical seal and Separator.

#### (j) Sports materials:

Spikes, Golf articles, Tennis racket, Fishing article, Mountain-climbing goods, Ski goods and Badminton racket.

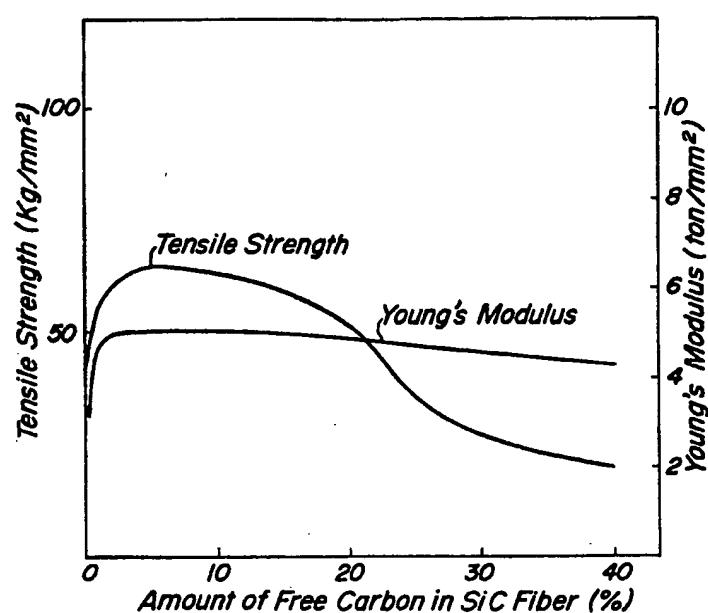
- (k) Ship and aircraft materials:  
Engine, Construction materials, Screw and Wing.
- (l) Electrical materials:  
Transmission cable, Condenser, and Telegraph Pole.
- 5 (m) Architecture materials:  
Window sash, Construction materials and so on.
- (n) Agricultural machines, Atomic implements, Nuclear fussion furnace material, Sun heat utilizing material, Medical instruments, Cycle materials, Valve, Valve seat, Ring, Rod, Disc, Liner, Sand transport pump parts, Machine parts for treating dust, Die and nozzle for extrusion or injection of plastics and Reflection mirror.
- 10 WHAT WE CLAIM IS:
1. A composite material comprising an aluminum, aluminum alloy or a magnesium alloy having embedded therein reinforcing continuous silicon carbide fibers as hereinbefore defined, said material having been obtained by heating 30-80% by volume of aluminum, aluminum alloy or magnesium alloy matrix and 20 to 70% by volume of the continuous silicon carbide fibers containing 0.01-40% by weight of free carbon at a temperature which melts the aluminum, aluminum alloy or magnesium alloy.
  2. Composite material as claimed in claim 1, wherein said aluminum alloy consists of aluminum and at least one of titanium, chromium, silicon, manganese and calcium.
  - 20 3. Composite material as claimed in claim 1, wherein said magnesium alloy consists of magnesium and at least one of aluminum, manganese, zirconium, calcium, silicon, and yttrium.
  4. A method for producing composite material which comprises contacting 20 to 70% by volume of continuous silicon carbide fibers containing 0.01-40% by weight of free carbon with 30-80% by volume of fused aluminum matrix, fused aluminum alloy matrix or fused magnesium alloy matrix so that the free carbon in the silicon carbide fibers reacts with the metal to form carbides of the metal.
  - 25 5. A method as claimed in claim 4, wherein the aluminum alloy contains one or more of silicon, manganese, chromium, titanium and calcium.
  - 30 6. A method as claimed in claim 4, wherein the magnesium alloy contains one or more of aluminum, manganese, zirconium, silicon calcium or yttrium.
  7. A method as claimed in any of claims 4 to 6 in which the continuous silicon carbide fibers, are precoated with at least one metal or with a ceramic material.
  - 35 8. A method as claimed in claim 7, wherein the coating metal is B, Mn, Mo, Al, W, Si, Cr, Ca, Ce, V, U, Th, Nb, Ta, Ti, Zr or Hf.
  9. A method as claimed in claim 7, wherein the coating metal is Be, Mg, Fe, Co, Ni, Cu, Zn, Ge, Pd, Ag, Cd, Sn, Sb, Pt, Au, Pb or an alloy consisting of at least two of the above described metals.
  - 40 10. A method as claimed in claim 7, wherein said ceramic material is Mgo, Al<sub>2</sub>O<sub>3</sub>, TiO<sub>2</sub>, ZnO, AlN, Mg<sub>2</sub>N<sub>2</sub>, Si<sub>3</sub>N<sub>4</sub>, TiN, ZrN, TiC, ZrC or WC.
  11. A method as claimed in claim 7, wherein a thickness of the metal or ceramic coating is 100-2000Å.
  - 45 12. A method as claimed in claim 7, wherein the coating is carried out by a chemical vapor deposition process, a flame fusion spray coating process, a spattering coating process, a vacuum evaporating coating process, an electroplating process, a powder baking process or an electroless plating process.
  13. A method for producing a composite material as claimed in claim 4 substantially as hereinbefore described.
  - 50 14. A composite material as claimed in claim 1 substantially as hereinbefore described.
  15. A composite material whenever produced by the method claimed in any one of claims 4 to 13.
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FIG.-I



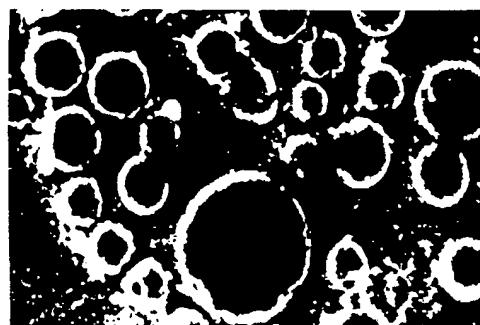
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*FIG.2*



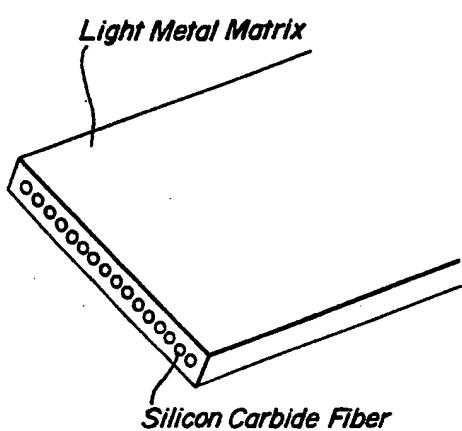
*FIG.4*



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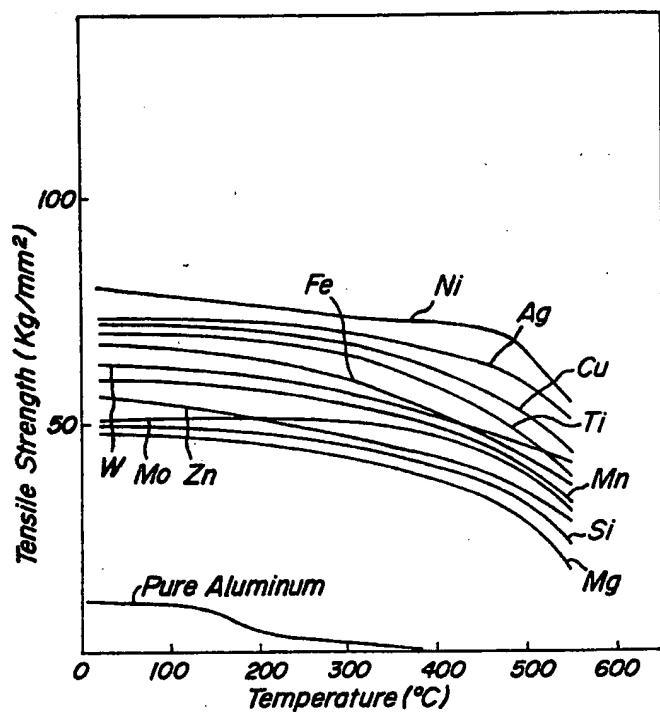
*FIG.3*



1572460 COMPLETE SPECIFICATION

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FIG.5



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